Salt Preparation and Reconditioning for Accelerator Driven Subcritical Fission in Molten Salt (ADSMS)

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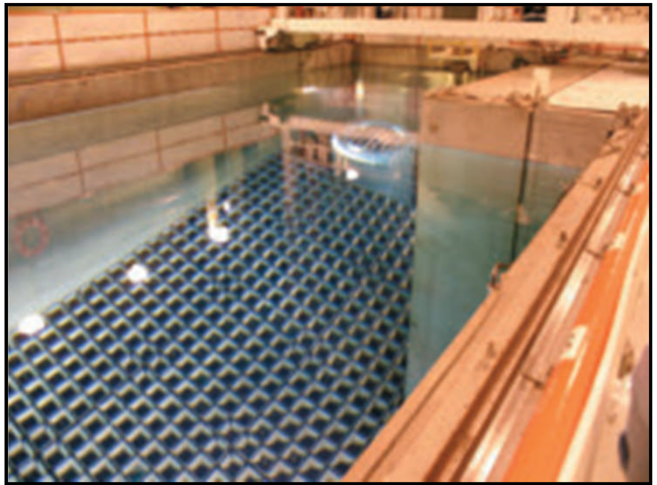
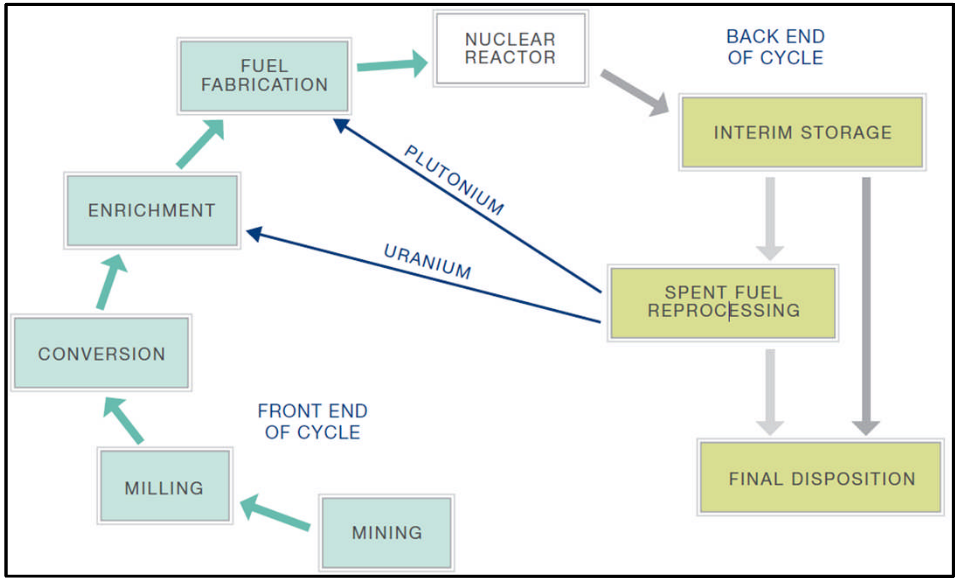
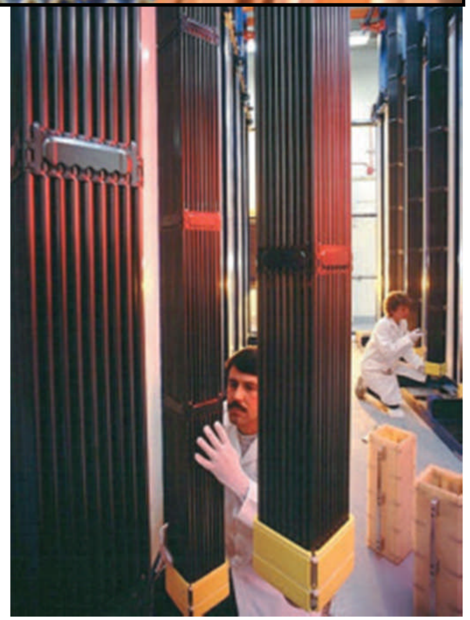
Used Nuclear Fuel (UNF) and The Fuel Cycle

- **UNF is stored on location in water ponds and dry casks.**
- **In this form it would remain hazardous for thousands of years.**
- **The only way to remove those hazards is to literally destroy them by fission for the transuranics (TRU).**



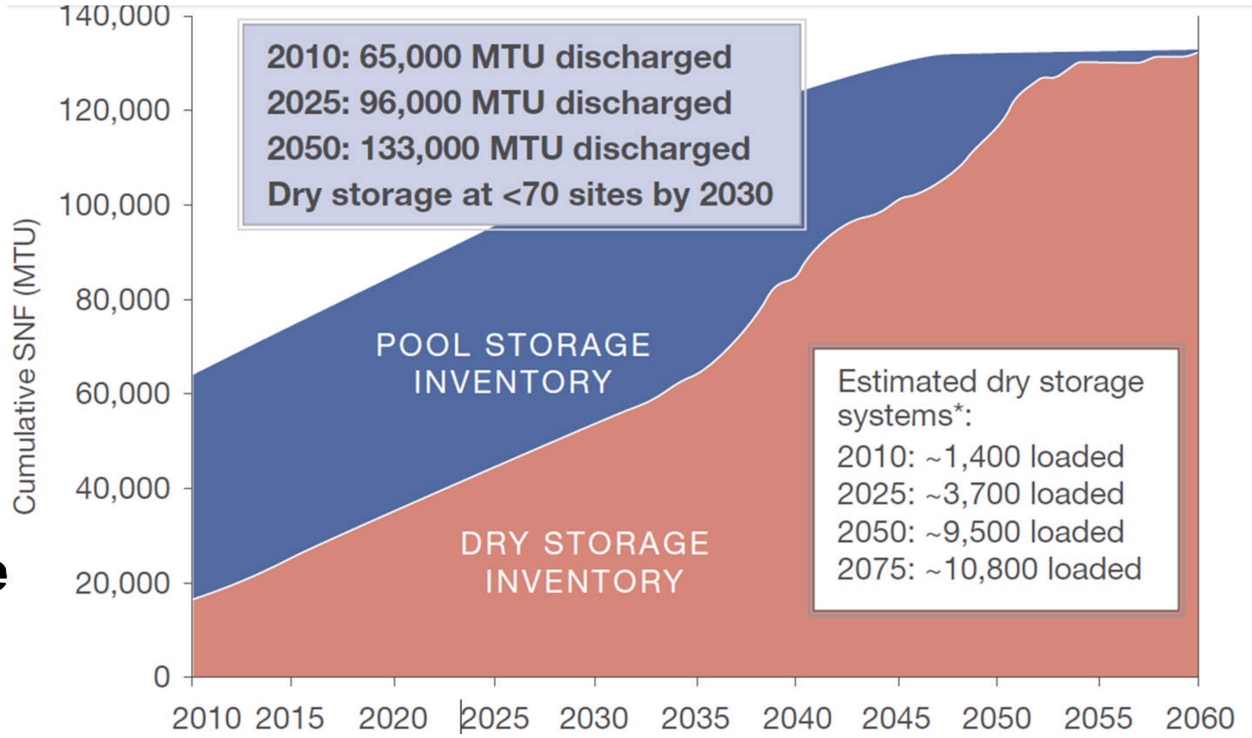




Reference: Blue Ribbon Commission on America's Nuclear Future, Jan 2012

A Call for Waste Disposal

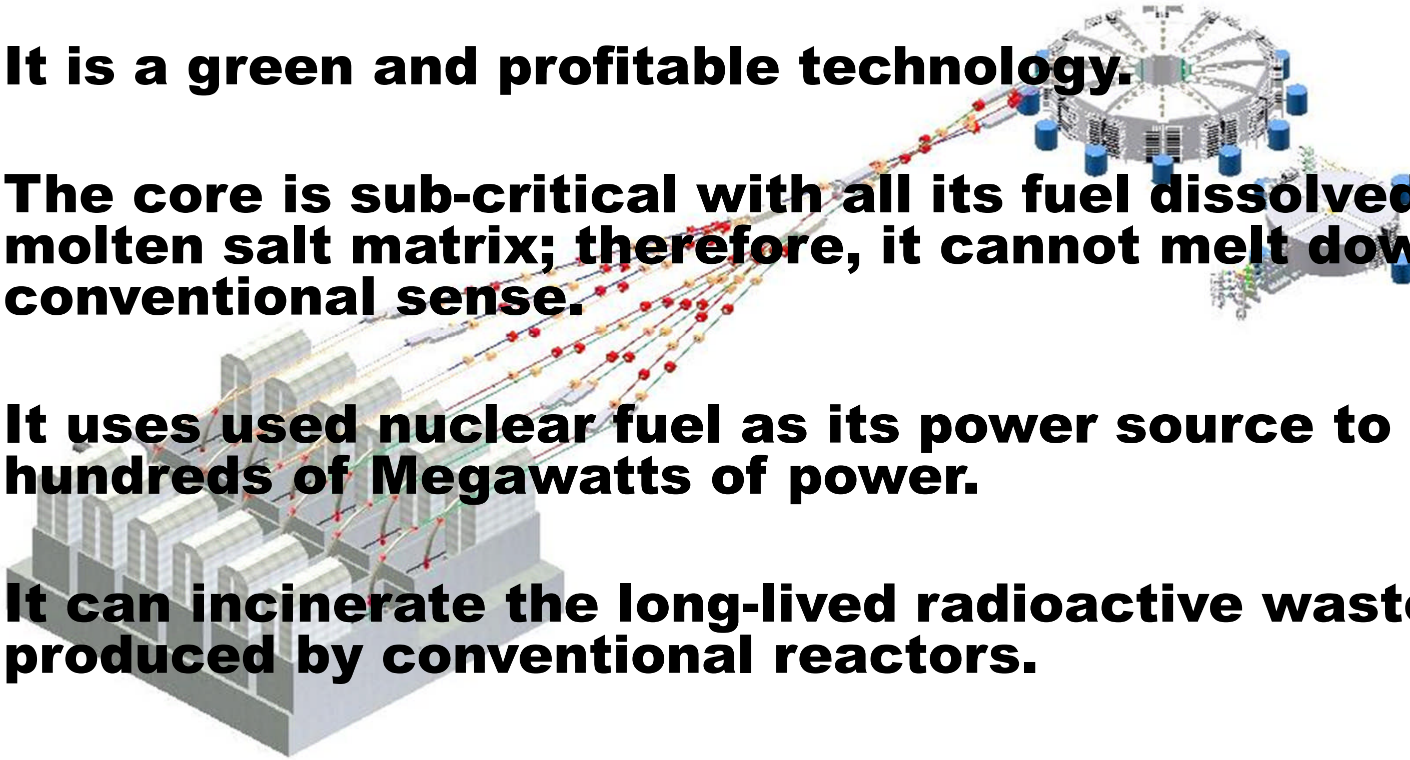
- **U.S. Used Nuclear Fuel Inventory:**
  - **Currently 67,000 Metric Tons of UNF in storage**
  - **Growth Rate of ~2000 Metric Tons per year**



Reference: Blue Ribbon Commission on America's Nuclear Future, Jan 2012

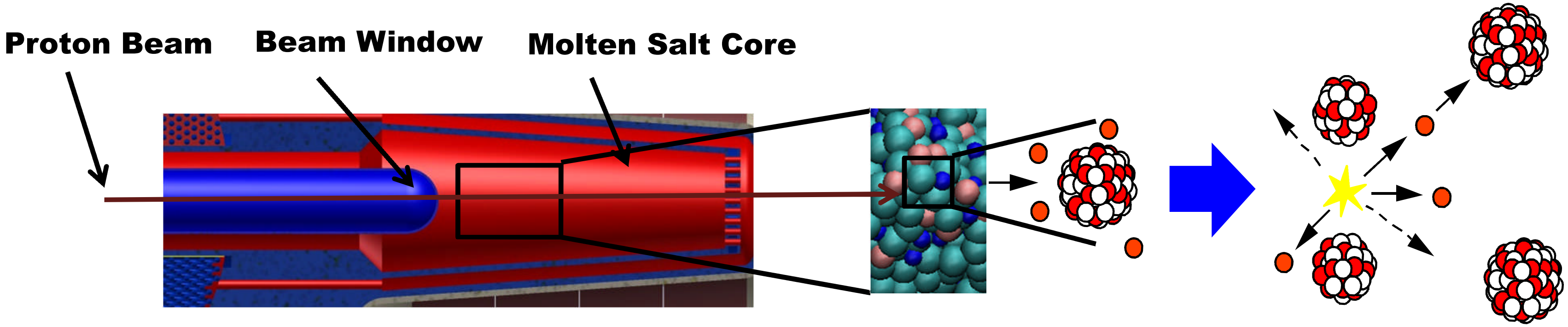
Benefits to ADSMS

- ADSMS can destroy the transuranics (TRU) at a rate equivalent to their production in conventional reactors.
- It is a green and profitable technology.
- The core is sub-critical with all its fuel dissolved in a molten salt matrix; therefore, it cannot melt down in a conventional sense.
- It uses used nuclear fuel as its power source to produce hundreds of Megawatts of power.
- It can incinerate the long-lived radioactive waste that is produced by conventional reactors.
- It can recycle used fuel into fertile fuel for conventional reactors.



Accelerator Driven Systems

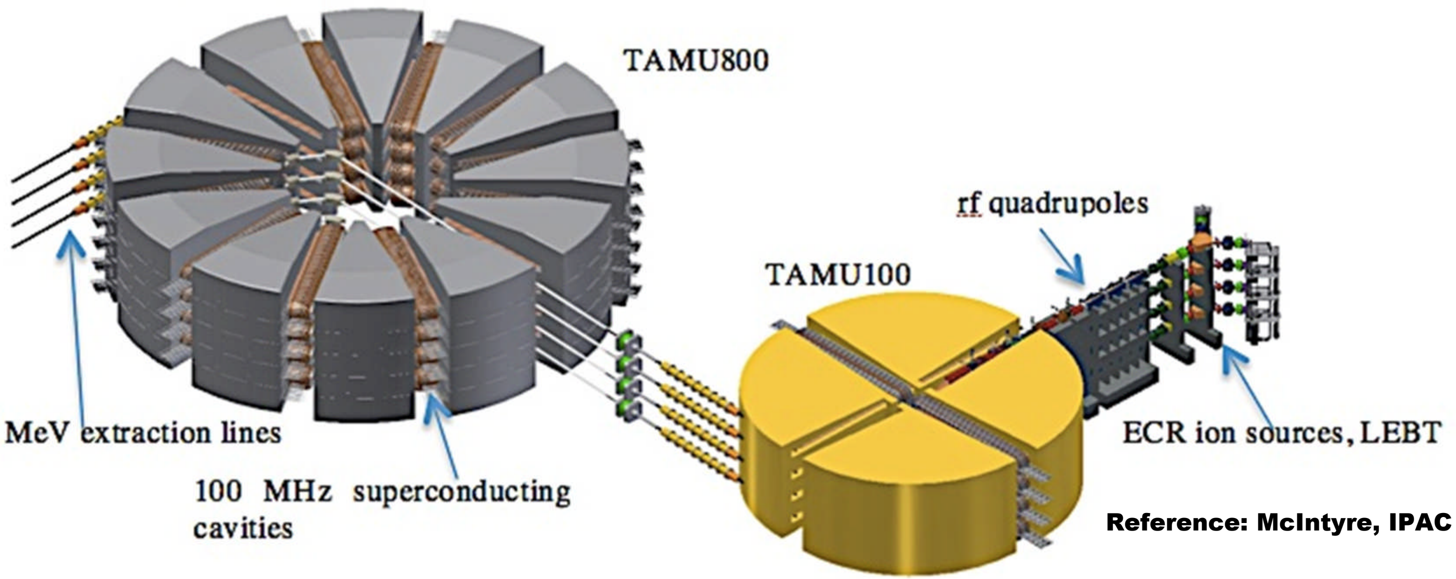
- **Why an accelerator?**
  - **An on-off switch to fission.**
  - **Capable of operating subcritical**
  - **Can run off of UNF**
- **Fission Driven by Spallation Neutrons**



- **Fast neutron source from 800 MeV Proton Beam**
- **Flux Coupled Stack of Isochronous Cyclotrons**
  - **1 footprint**
  - **Redundancy → reliability**
  - **Sustainable driver for fission cores**

Accelerator Complex

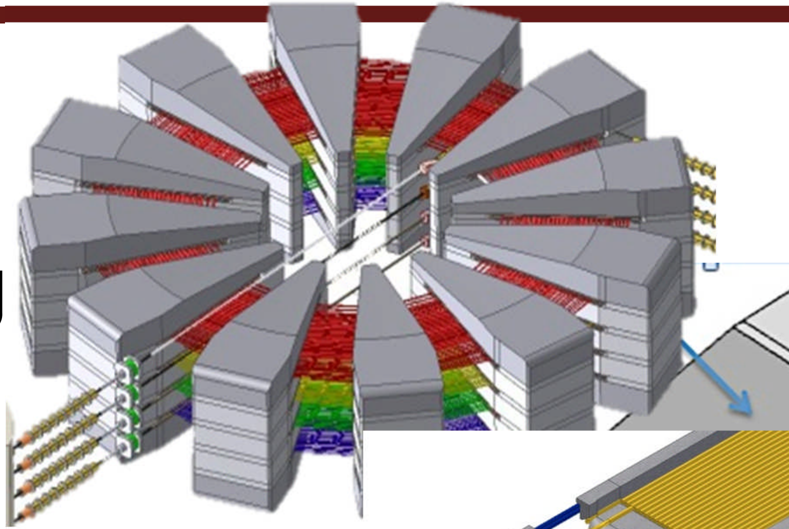
- **Injector**
  - **2.5 MeV proton beam is fed to the TAMU 100.**
- **TAMU 100**
  - **Produces a 100 MeV proton beam is fed to the TAMU 800.**
- **TAMU 800**
  - **10 mA beam from each cyclotron is chopped at injection and then separated into 3 beams after extraction from TAMU 800.**
  - **The 4-stack of cyclotrons can produce 12 2.7 MW beams.**
- **12 beam lines**
  - **Each beam can drive an 65 MW_{th} ADS molten salt core.**

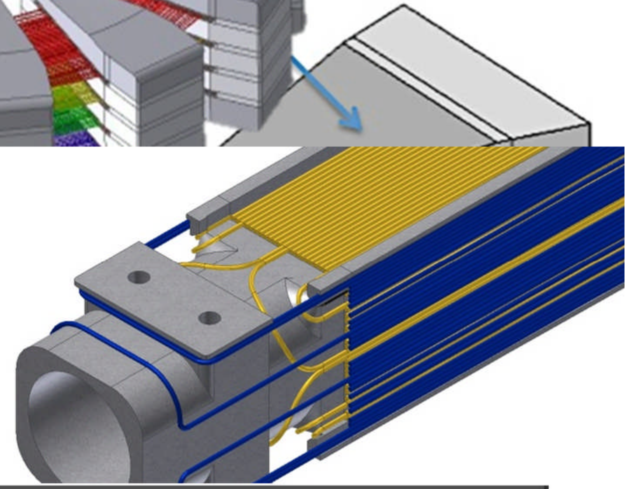


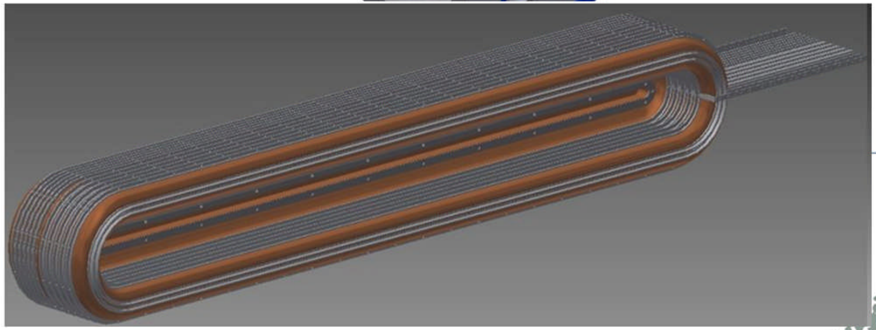
Reference: McIntyre, IPAC 2012, Poster Session

Accelerator Technology Advances

- **Flux Coupled Stack**
- **Strong Focusing Cyclotron**
  - **Quadruple focusing channels at the magnet pole faces**
  - **Novel Design for Superconducting RF Cavities**

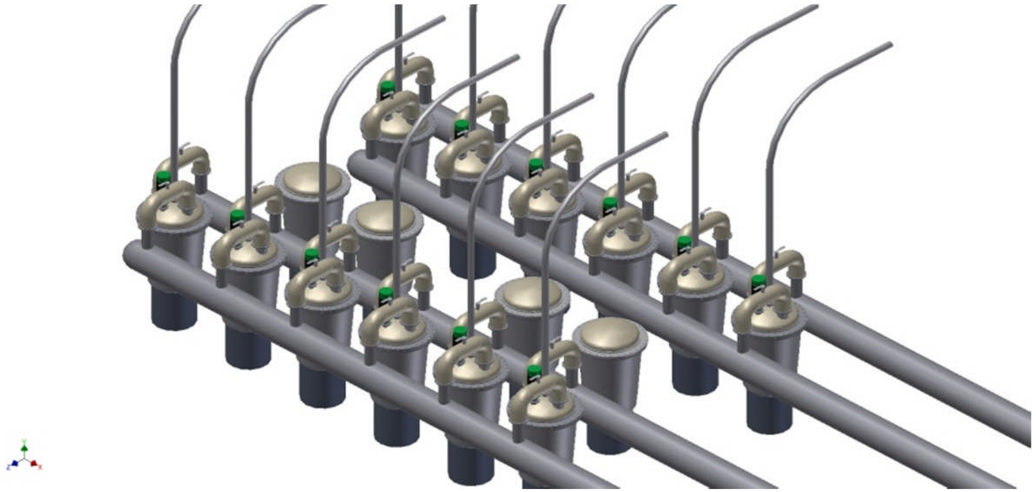


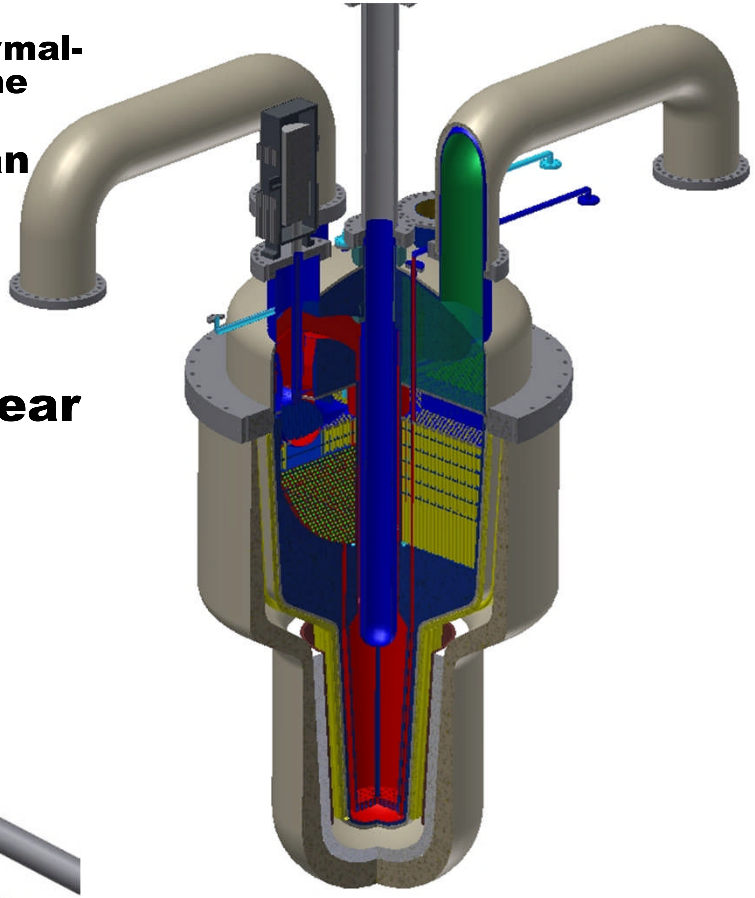




Molten Salt Fission Pot

- **12 Molten Salt Pots**
  - **65 MW_{TH} Each**
  - **Produce 300MW_{E} together**
    - **Energy gain of 5.3 taking into account thermal-electric conversion and the efficiency of the accelerators**
  - **Together they can burn the contents of an AP1000**
- **NaCl-Based Molten Salt**
  - **NaCl – 69%**
  - **TRU (0.878) +Lanthanides (0.122) = 31%**
- **Salt processing and refueling every year**





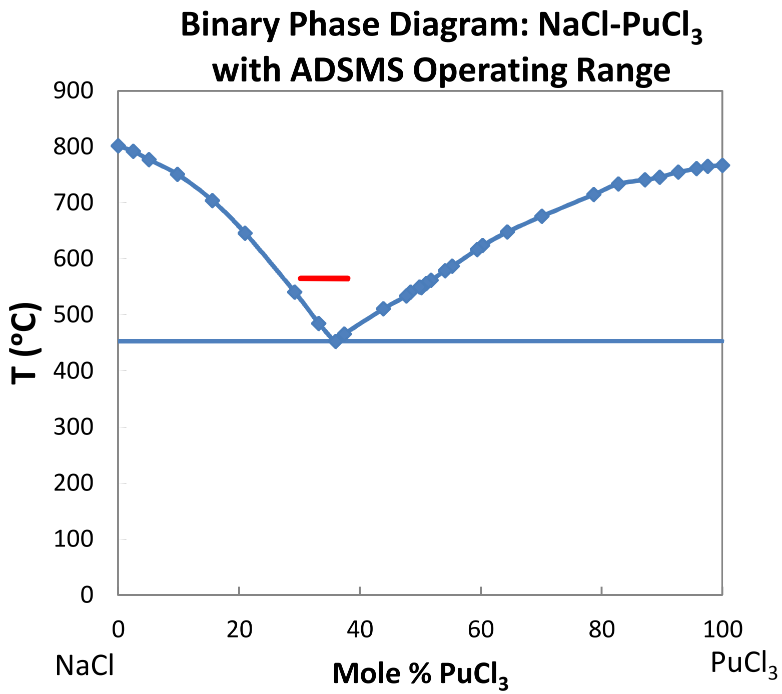
Chloride Molten Salt System

- **Why Chlorides?**
  - **High actinide solubility**
  - **Compatibility with very fast neutronics**
  - **Stable operating temperature range ~500-700 °C**
  - **Low chemical corrosiveness**
- **Why not Chlorides?**
  - **Cl^{36} Production**
    - ***Solution: Isotope Separation (USEC)***
  - **Complex chemistry**
    - **Multiple oxidation states**
  - **Limited Experimental Data**

	Fluorides	Chlorides
Actinide Solubility at Acceptable Temperature	Low	High
Vapor Pressure	Very Low	Low
Operating Temperature	700 °C+	500 °C+
Electronegativity (Pauling Scale)	3.98	3.16
Number of Oxidation States	1	8
Number of Stable Actinide Ionization States	Several	1-2

Chloride Molten Salt Systems

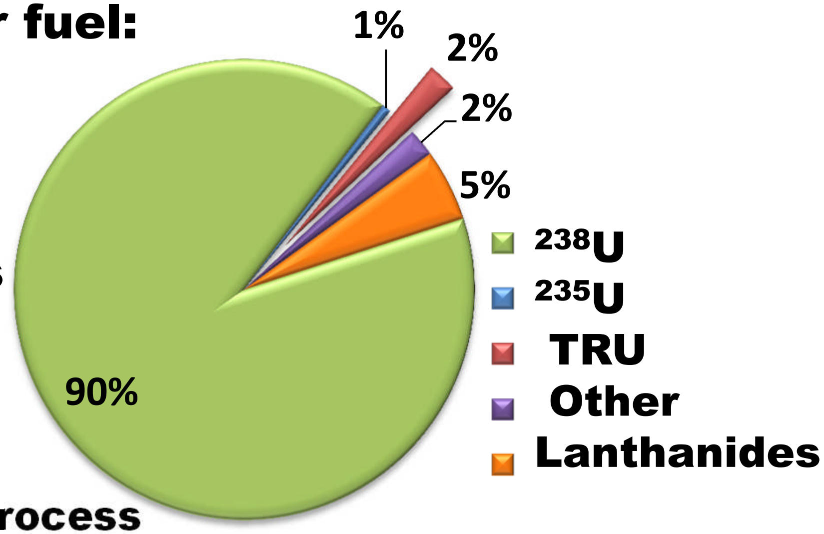
- **LiCl-KCl**
  - **Pyroprocessing advantages**
  - **Wealth of available data**
  - **Secondary Salt**
  - **Not suitable for primary salt**
- **KCl**
  - **n,α reaction, ^{36}Cl production**
- **$MgCl_2$**
- **NaCl**
  - **High actinide solubility**
  - **Well modeled**
  - **Primary Salt Choice**

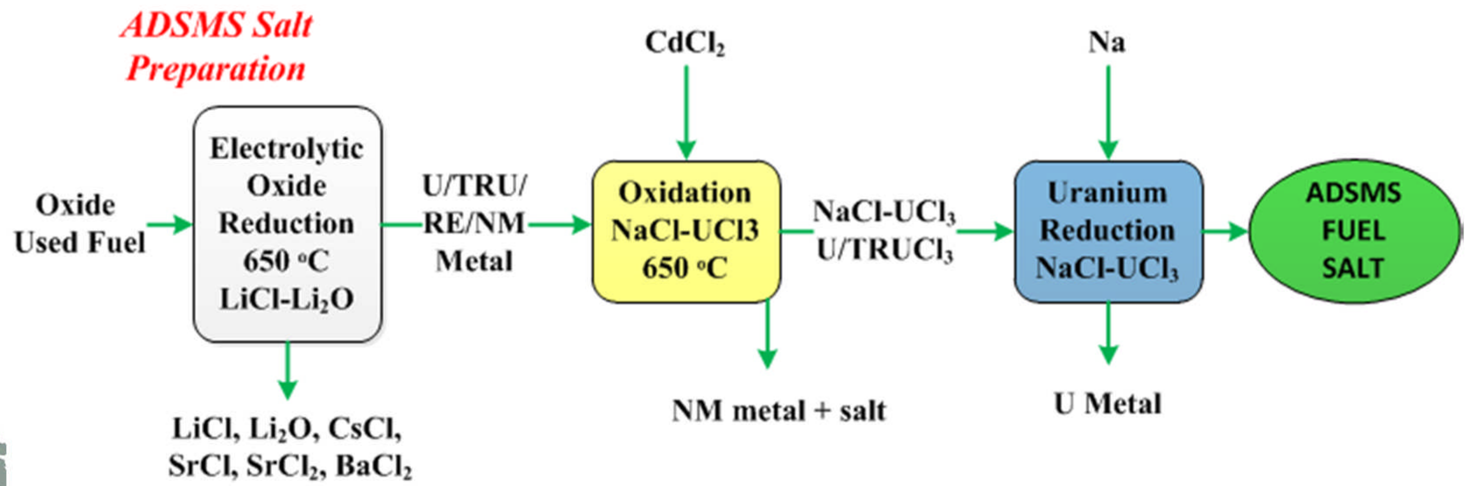


Reference: Bjorklund, *et al*, 1959.

Fuel Salt Preparation

- **Preparation of Oxide Spent Nuclear fuel:**
  - **Electrolytic Oxide Reduction**
  - **Rare Earth and Actinide Oxidation**
  - **Uranium Reduction**
- **Avoids typical proliferation hazards**
  - **Non-aqueous**
  - **1 Pot Process**
  - **No segregation of Pu**
  - **Radioactive lanthanides remain with the minor actinides throughout the process**

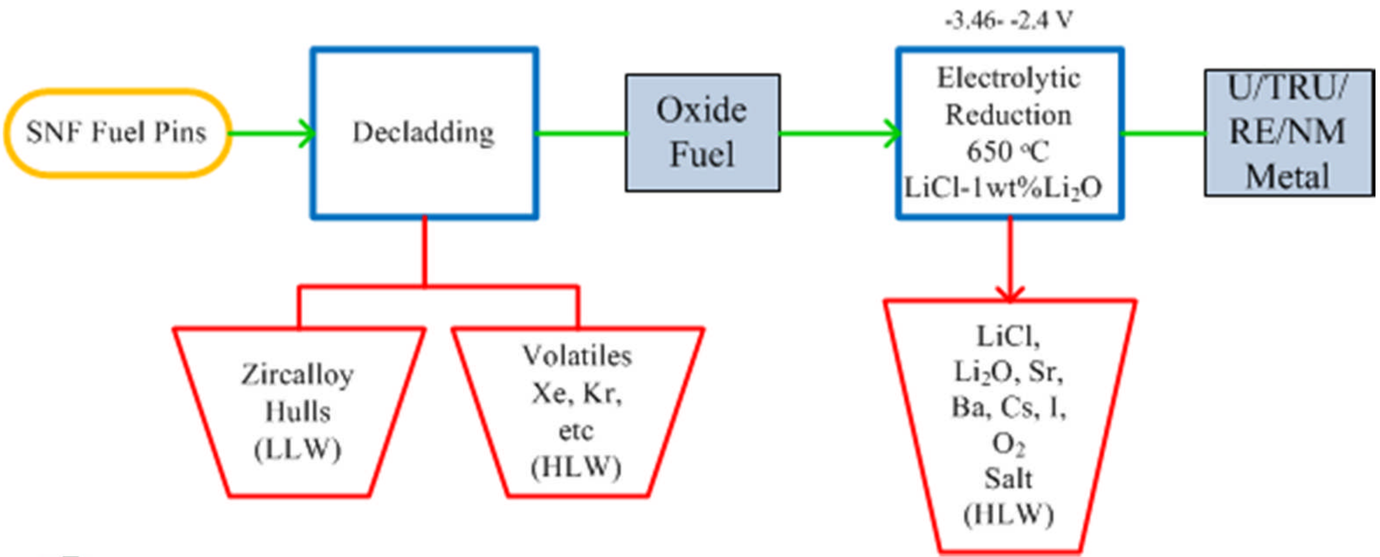




Electrolytic Reduction

- **Developed at ANL and INL**
  - **LiCl-1wt% Li_2O**
  - **Uranium, TRU, Noble Metals, and Rare Earth Reduction in Cathode Basket**
  - **>90% Reduction of TRU**

Oxide Fuel before and after Voloxidation

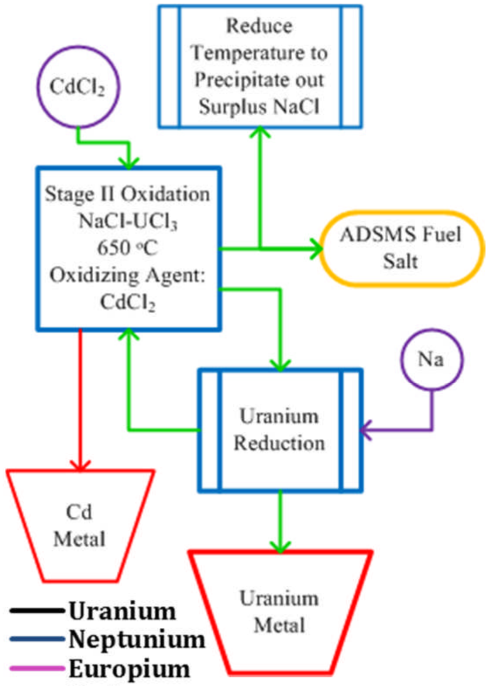


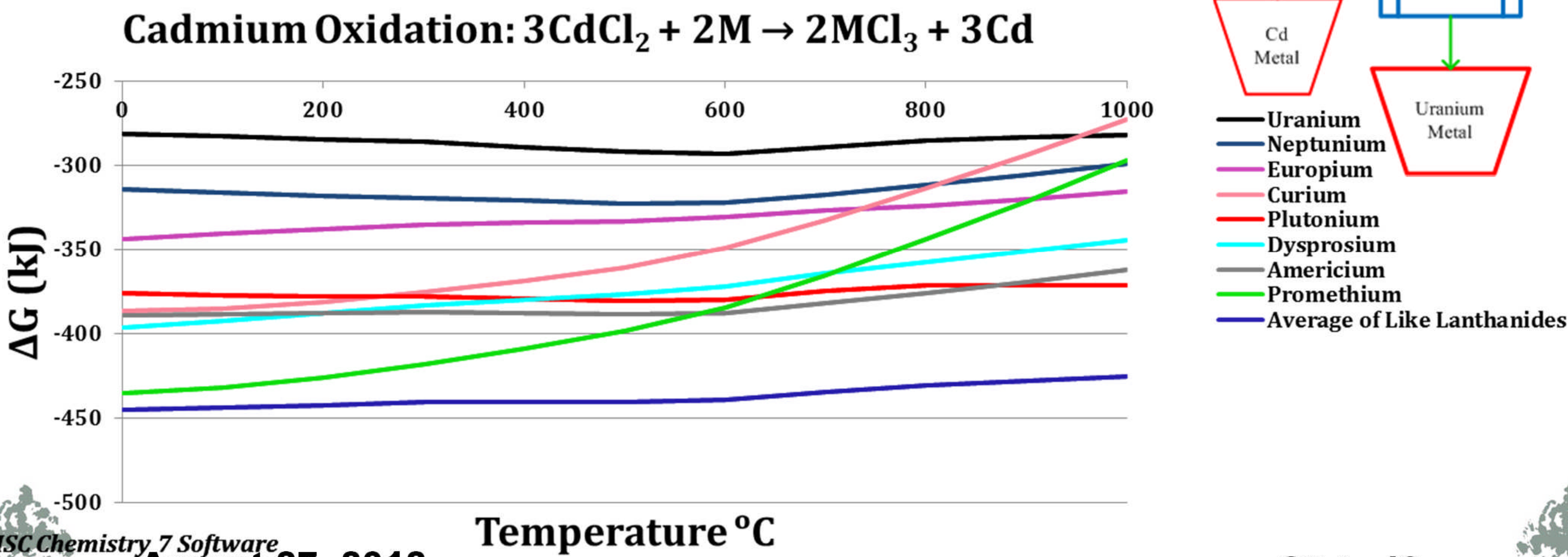
Sintered Oxide Fuel

Post Electrolytic Reduction of Oxide Fuel

Heavy Metal Oxidation

- $CdCl_2$ Oxidation
- Lanthanides Oxidize first
- Uranium oxidizes last of the Actinides
- Noble Metals are left as waste product
- UCl_3 can provide lower melt temp for operation.

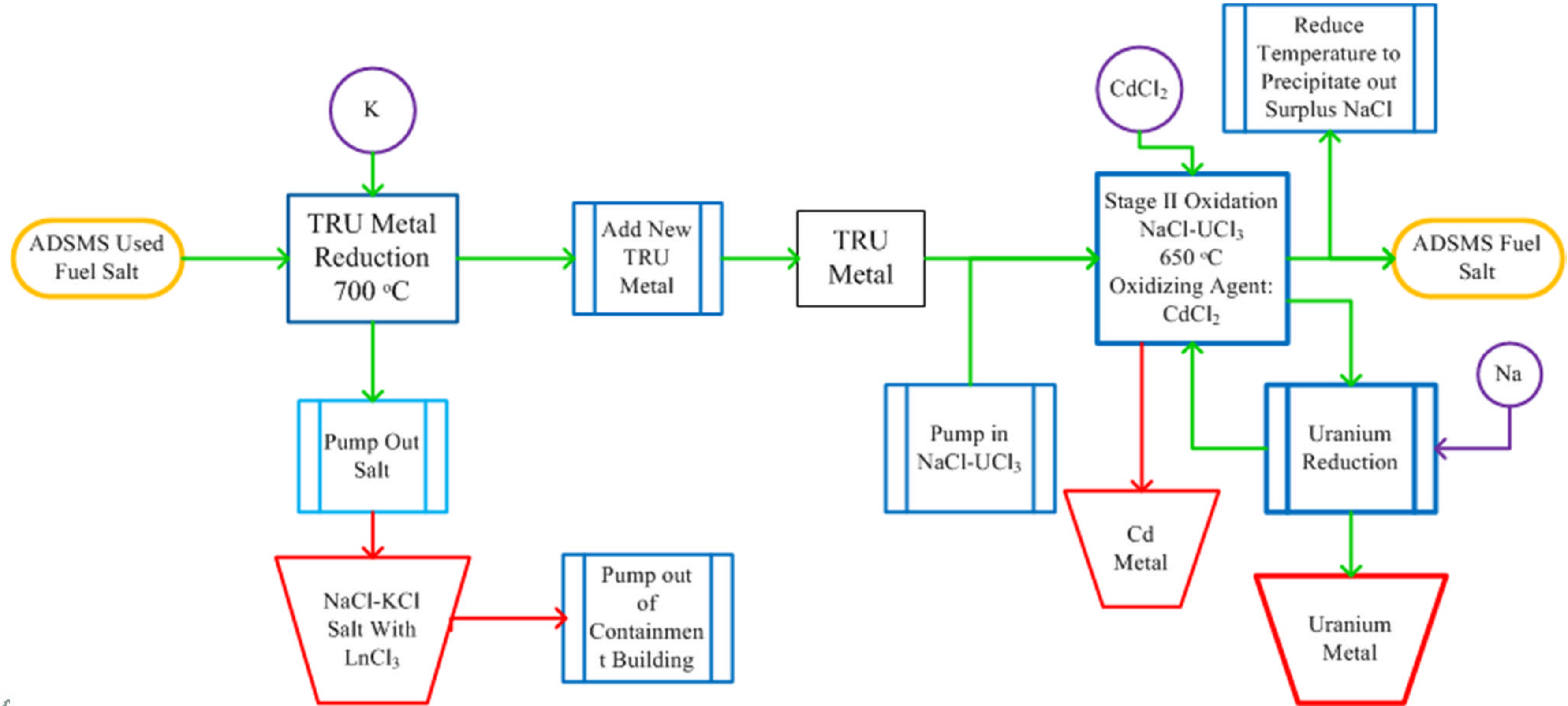




HSC Chemistry 7 Software

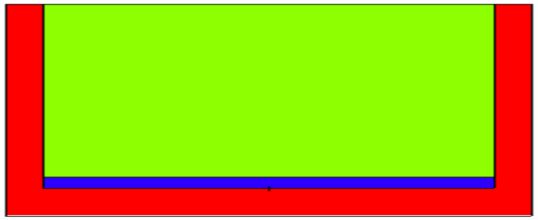
Molten Salt Reconditioning

- **Similar steps as Salt Preparation**
- **Potassium reduction helps to keep lower operating temperature and avoid salt freezing**
- **Criticality concerns mitigated via batch processing**

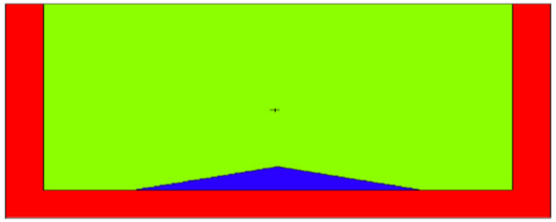


Criticality Concerns During Reconditioning

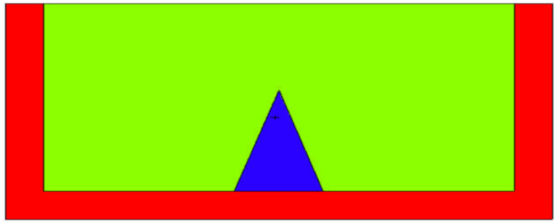
- During reconditioning, TRU is reduced into metal form from the molten salt.
- Criticality calculations were performed with the Monte Carlo code MCNPX for several vessel designs and deposition scenarios.
- Critical mass calculations are sensitive to:
  - Deposition Geometry
    - Dendrite formation or preferred TRU deposition decreases the critical mass
  - Vessel Design
    - Bucket Shape
    - Infinite lattice geometry- series of individual cells can be modeled and share neutrons
  - Absorber width and placement
    - Hafnium wall thickness has an exponential on criticality- diminishing returns



Uniform deposition
Critical mass: 168.3 kg



Cone half-angle 75°
Critical mass: 35.1 kg



Cone half-angle 15°
Critical mass: 14.4 kg

Conclusions

- **Proposing a power producing solution to the nuclear waste problem.**
- **Making *innovative* advances in accelerator technology.**
- **Actively seeking**
  - **Experimental and modeling support, suggestions, and lessons learned and**
  - **Collaborations to assist with actinide salt experiments, corrosion, and radiation damage testing.**

The ADSMS Collaboration

Texas A&M University:

Physics:
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Karie Badgley
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Justin Comeaux
Tim Elliott
James Gerity
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Joshua Kellams
Al McInturff
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Mechanical Engineering:
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Chemistry:
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Deepak Raparia
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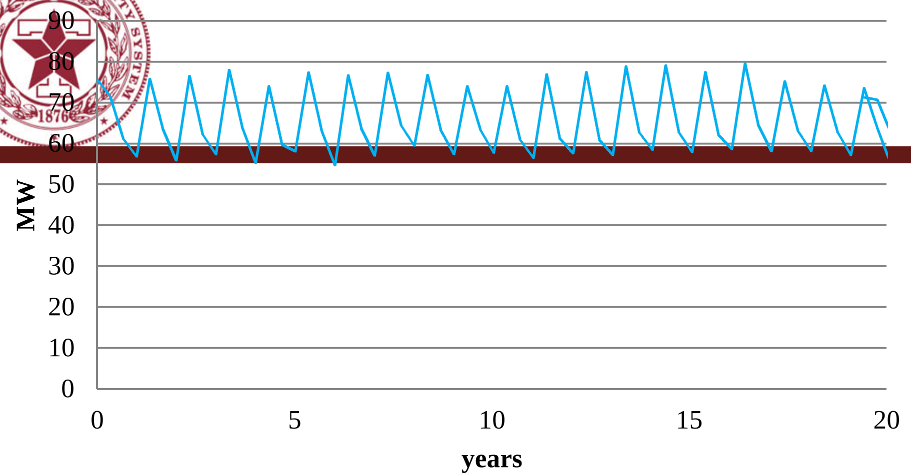
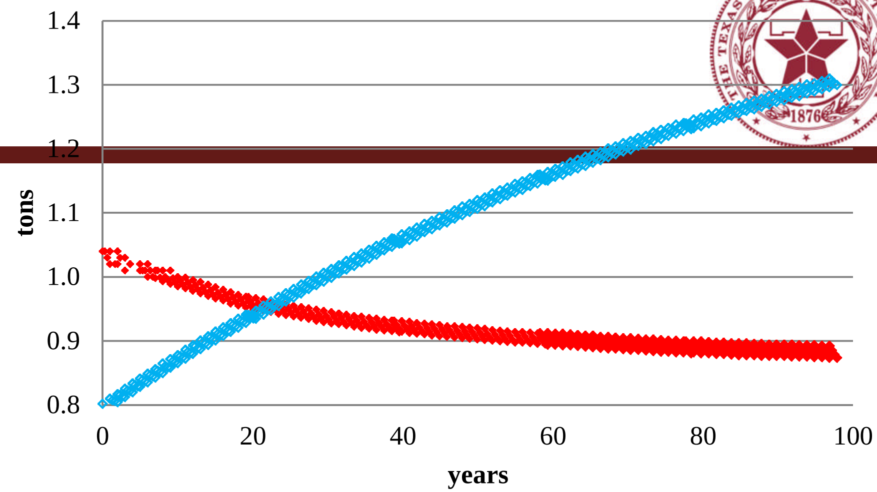
Questions?

Thank you for your attention!

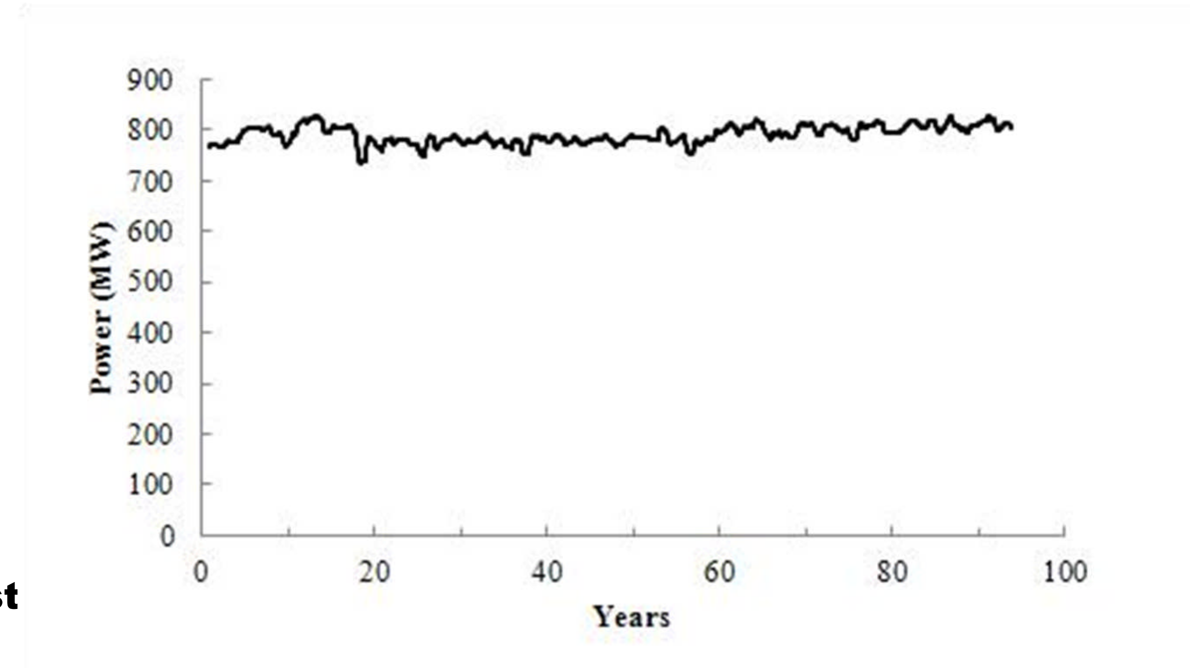
W_{th} one core



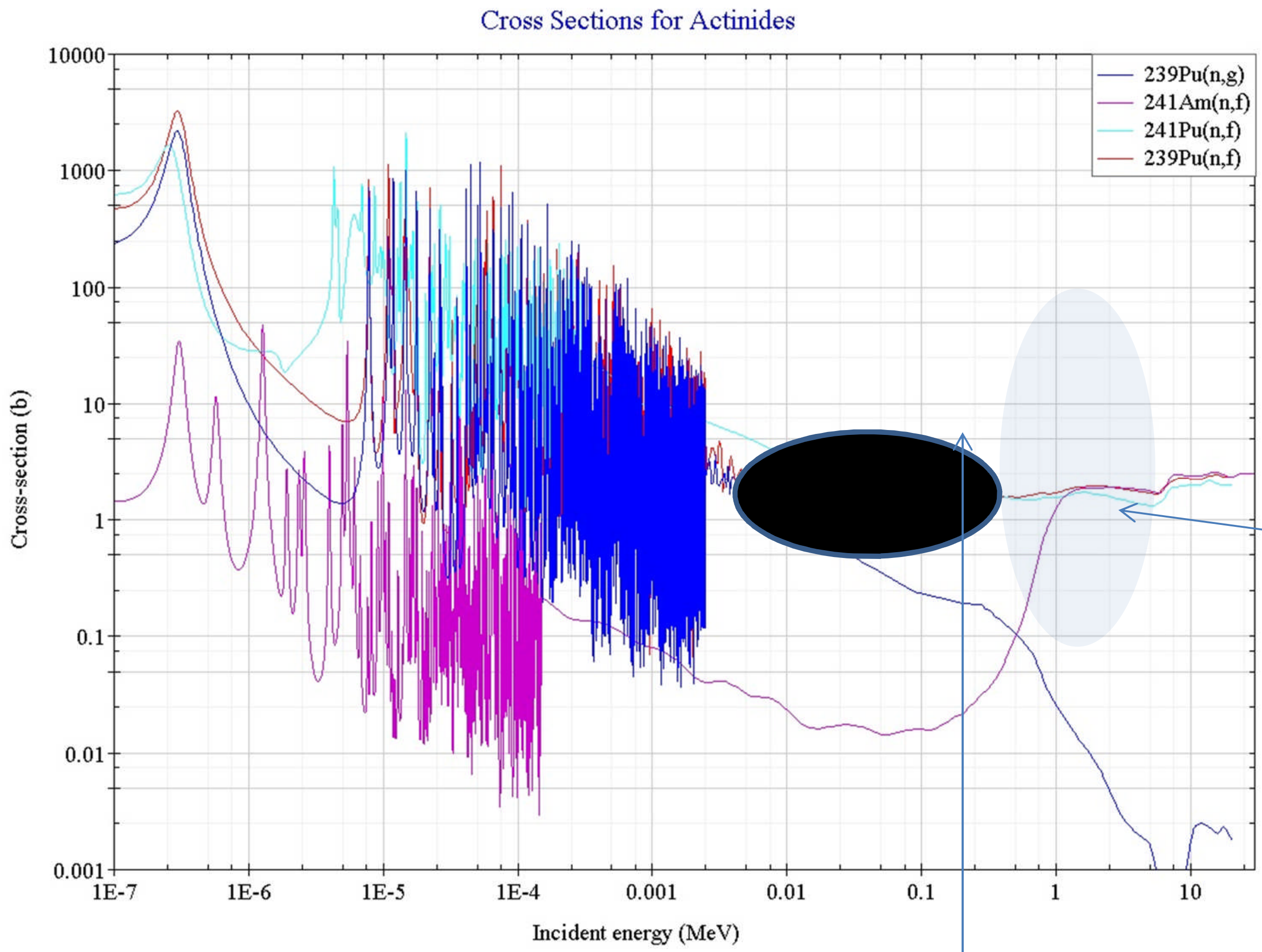
^{239}Pu and TRU' inventories 1 core



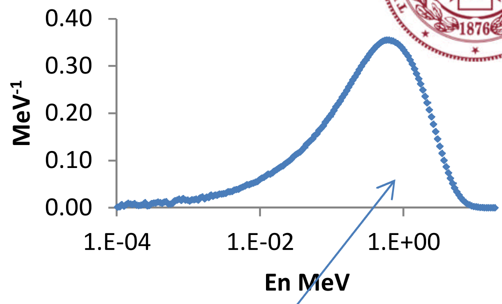
W_{th} from 12 core ADSMS



August



Fission Neutron Spectrum from ^{239}Pu

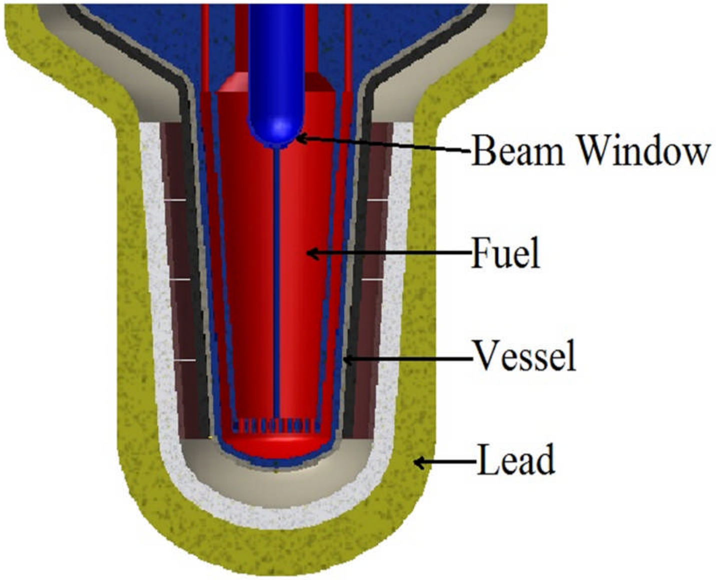


Neutrons are born in this region. All Actinides can fission at rates proportional to their fraction in the fuel

If they do give any fission (no capture), they scattering in the medium they loose their energy, move this way on the plot. Only ^{239}Pu can fission !!!

Outside second region capture becomes also important and as a result one starts to breed back minor actinides. We need ultra fast neutrons in the core, smaller core!!!!

W_{th}, MW	800
Proton Beam	4, 800MeV, 10mA
Total flux, $10^{15}cm^{-2}s^{-1}$	1.3
Fast fraction, n_f	28%
k_{eff}	0.96
Burn Up (GWd/tHM)	500 (100y)
T_{melt}, ^oC	515
Composition	31%(TRU+La)Cl_3 69%NaCl
Density, g/cc	3.142
Total load, t	22.2 (56 in 100y) Actinide
Power density, W/cc	165W
T_{in}/T_{out}, ^oC	565/575

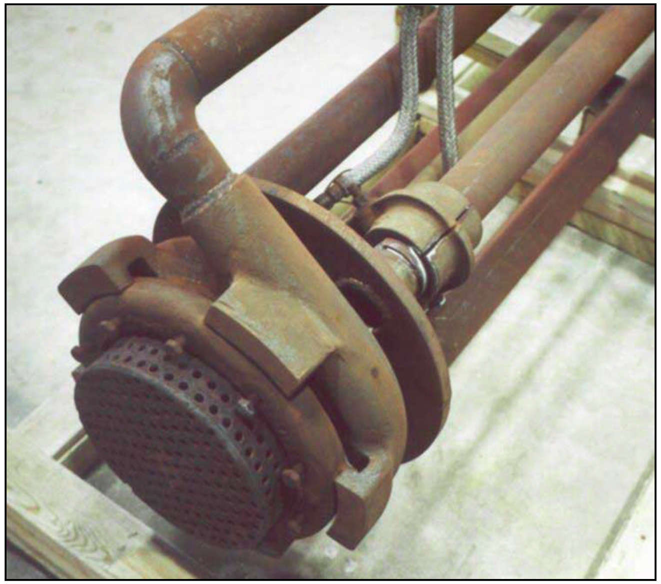


Main vessel dimensions	
Inner radius	0.32 m
Height	1.5 m
Outer radius of downflow tube	0.27 m
Thickness of downflow tube	0.0254 m
Manifold plate thickness	0.01 m
Wall material/Thickness	
Ni	0.00635 m
Hastelloy-N	0.0127 m
HT-9	0.0254 m
Ni Beam line and beam "windows"	
OD	0.26 m
Beam window shape	half sphere
Thickness	0.003 m
Absorber/Reflector	
B_4C	0.48 m radius, 0.1 m thick
Lead	0.65 m radius, 0.3 m thick

August 27, 2012

Objectives of Corrosion Testing

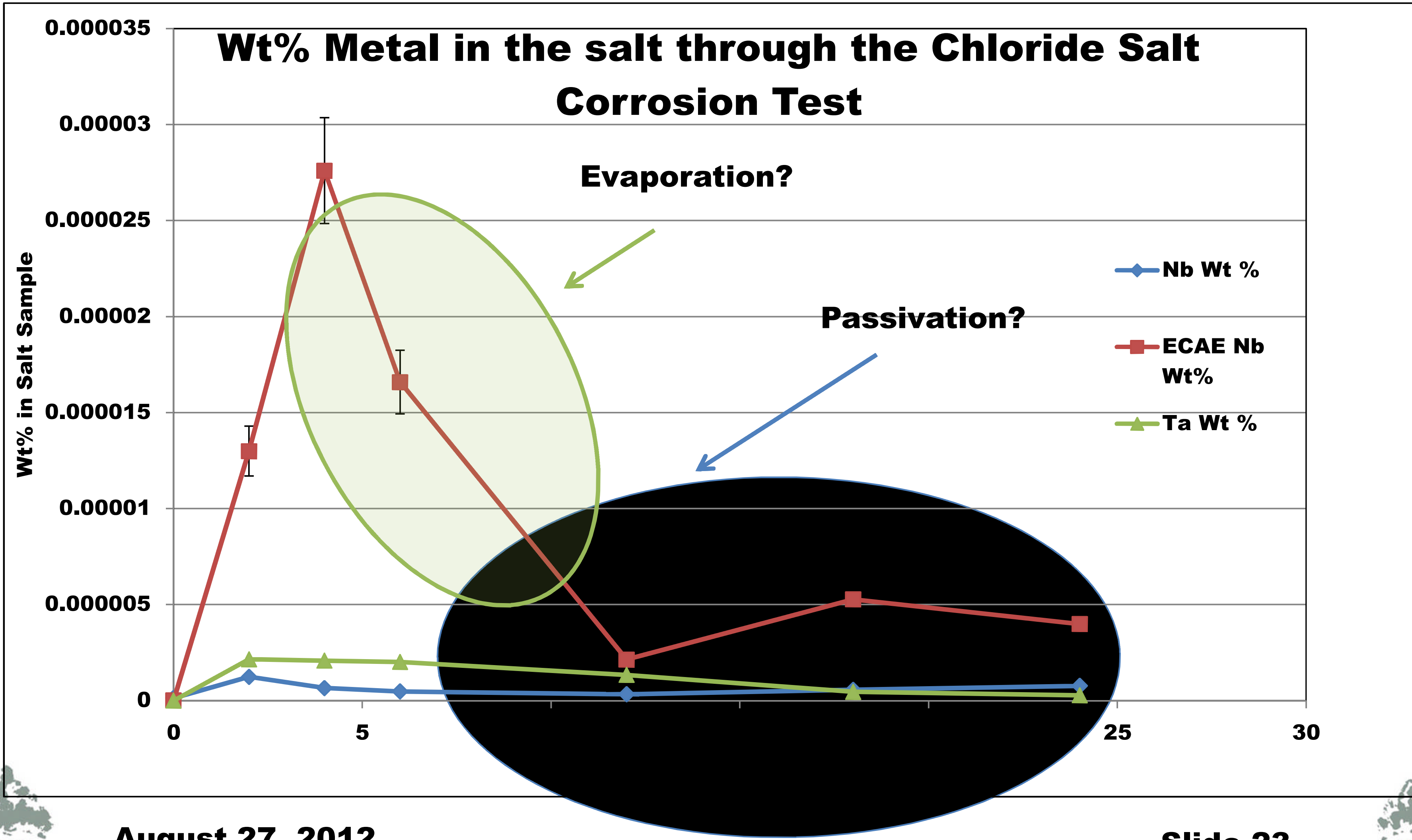
- Identify corrosion resistant materials in chloride molten salts
- Quantify the effect of various oxygen concentrations in the salt
  - Mimic realistic operating conditions
  - Simulate worst-case scenarios
- Control the environment and suppress corrosion
  - Monitor and stabilize redox potential in the salt
  - Cathodic Protection: Naval Ships and your Hot Water Heater
- Combine molten salt corrosion with neutron damage effects
  - Swelling
  - He Embrittlement



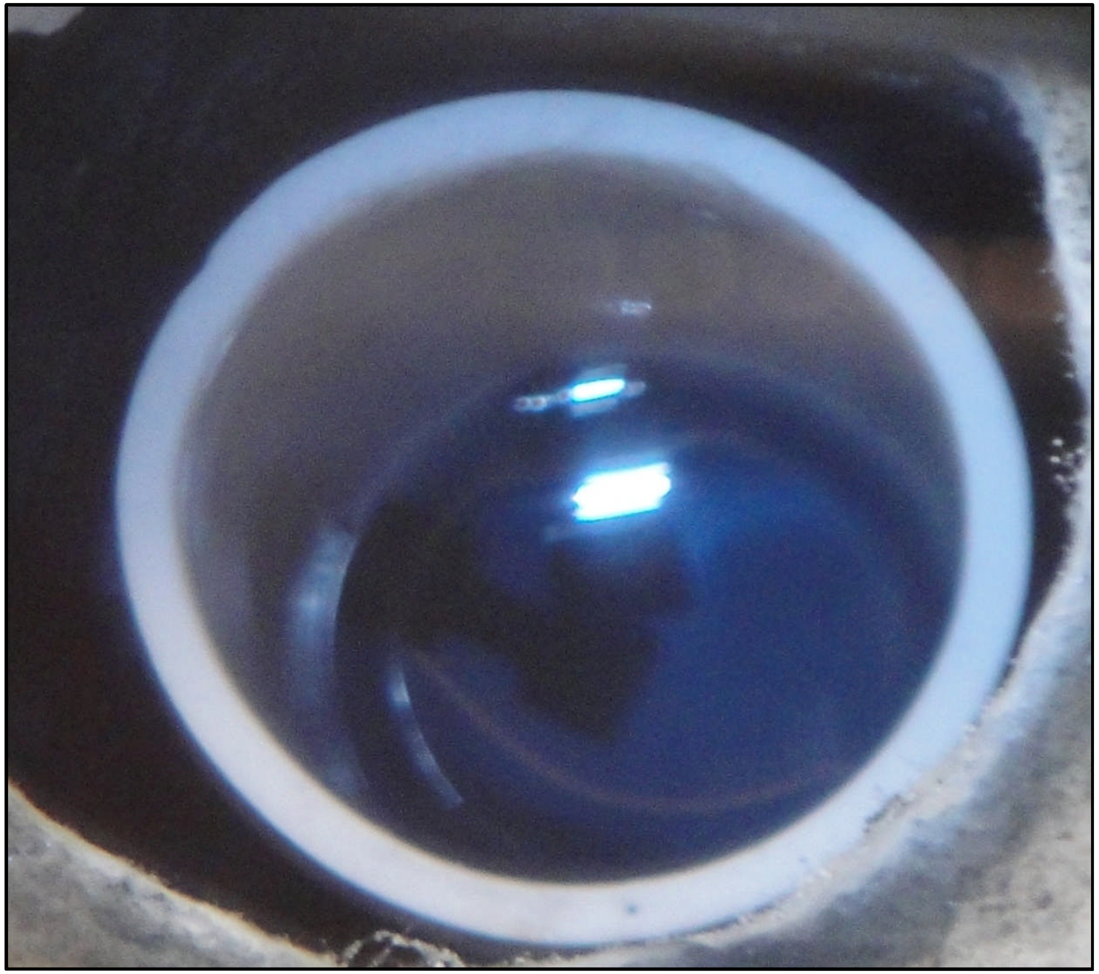
Centrifugal pump operated continuously for one year in molten salt at 500 C

Initial Mass Spectroscopy Data

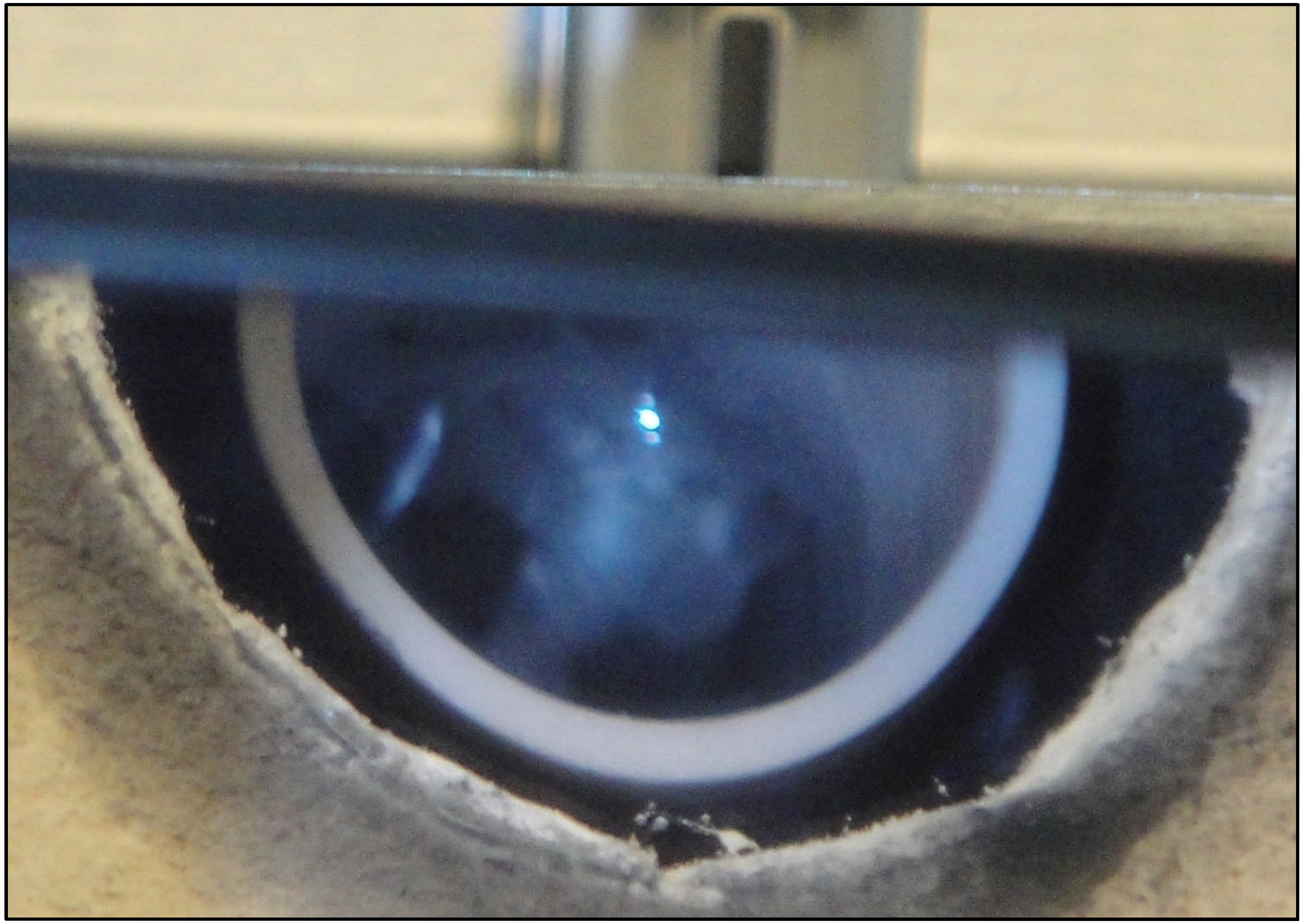
A clear trend was seen in the refractory material:



2) Blue salt: Zircaloy Exposure in Chloride Salt, t=2hr



3) Stainless Steel Exposure in Chloride Salt, t=6hrs



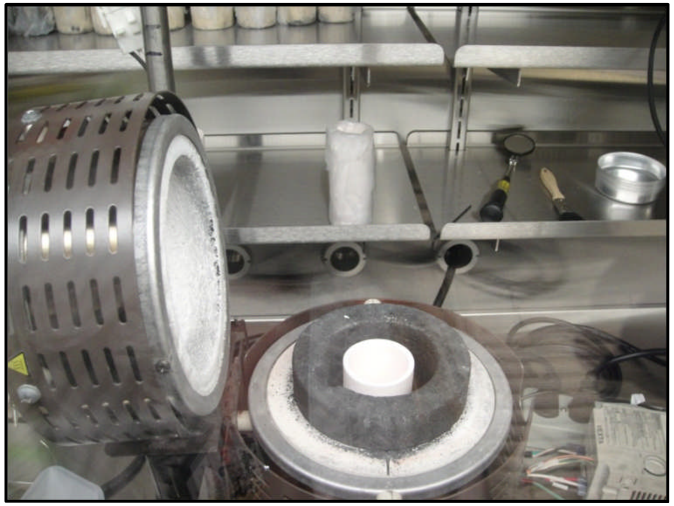
Initial Testing at the Center for Advanced Energy Studies

- **Baseline corrosion test:**
  - **LiCl-KCl and LiBr-KBr**
  - **11 different materials**
  - **Inert, argon glove box**
  - **24 hour experiments**
  - **Each experiment run at 700 °C**
- **Samples:**
  - **6 samples per metal**
    - **3 samples per salt compound**
      - **1 Control**
      - **2 Exposed to salt**
  - **All samples were etched and prepared in the same manner**
- **Salt and Materials Characterization**
  - **Inductively Coupled Plasma Mass Spectrometry**
  - **Scanning Electron Microscopy**
  - **Tension Test**

Sample Prep and Experimental Controls

- Etched according to ASM Handbook recommendations
- Rinsed with nano-pure water then dried using isopropyl alcohol and baked at 150 °C
- Exposed to salt for 24 hours
- Salt samples taken at 2, 4, 6, 12, 18, and 24 hours
- Same operating temperature for each experiment
- Eutectic salt cations remained consistent, i.e. LiBr-KBr and LiCl-KCl

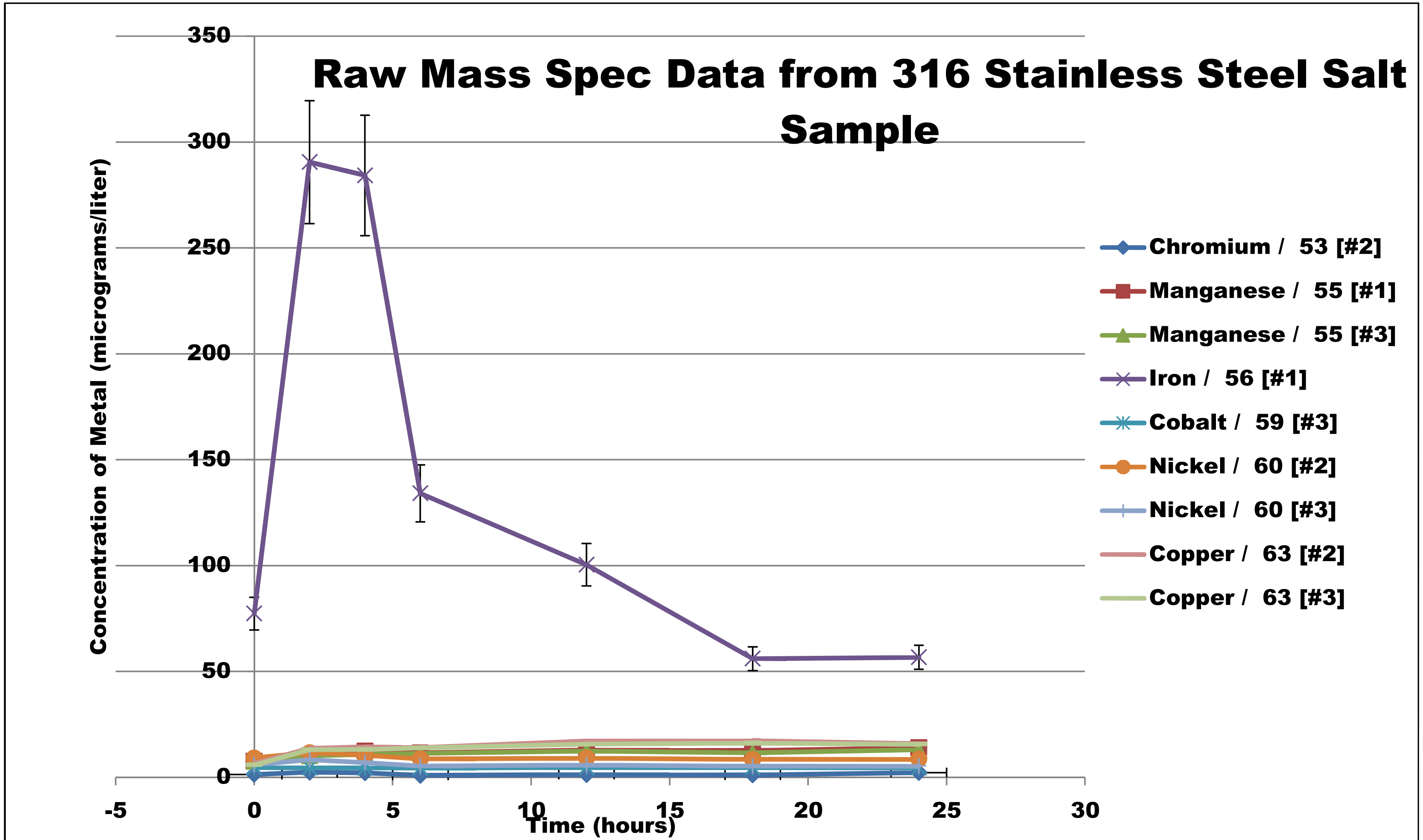
Zircaloy Samples before and after corrosion test



Future Corrosion Work

- **Continue salt analysis and evaluate precision of results**
- **Experiment with sample preparation techniques**
- **Next round of corrosion tests:**
  - 500 hours
  - Like materials for corrosion crucible and coupons
  - Nickel and Nickel Alloys in comparison with Stainless Steel
  - Gas monitoring of a closed system
- **Radiation damage of testing of Nickel and Nickel Alloys**
  - Simulate He embrittlement
  - Assess possibility to use Hastelloy-N or Pure Nickel for both vessel and heat exchanger tubing

Initial Mass Spectroscopy Data



We are seeking collaborators!

- **Neutronics**
- **Molten salt chemistry /corrosion**
- **High-power spallation targetry**
- **Beam dynamics – strong-focusing cyclotron**
- **Superconducting magnetics**
- **Superconducting RF**
- **Fuel cycle simulation/optimization**
- **Counter-flow heat exchanger**
- **Safety analysis, what-if scenarios**

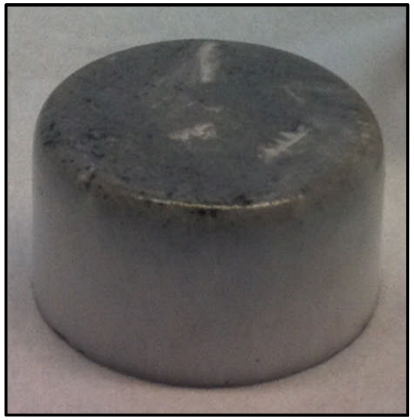
Please contact us - mcintyre@physics.tamu.edu

Metal Samples Tested:

- Niobium
- ECAE Niobium (Equal Channel Angular Extrusion)
- Tantalum
- Nickel
- Zircaloy (two types)
- 316 Stainless Steel
- Tungsten
- Hastelloy-N
- HT-9 Steel
- T91 Steel

Initial Observations Made

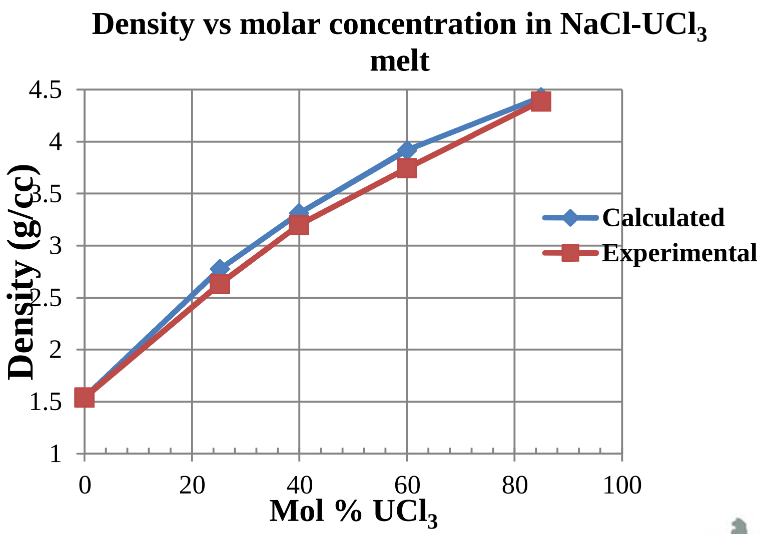
- Salt changed colors in a number of experiments
- Often times a metallic film appeared to cover the bottom of the crucible
- Vapor formed through the duration of the bromide studies
- Any wire used in early iterations of the experimental set up had clear vapor effects-color change and hardening



Tungsten Salt Column after Test (LiBr-KBr)

Modeling and Experimentation: Bridging the gap in data

- **Molecular Dynamics Modeling:**
  - **Ionic structure of a molten salt**
    - **Interaction Potentials: forces describe the salt's behavior**
      - **Intermolecular Bonds, Repulsive Forces, Dispersion, Electrostatics, and Polarization Effects**
  - **Solves the equations of motion at every time step**
- **With this program, we can calculate:**
  - ***Density: We have already implemented this in our neutronics calculations***
  - **Heat Capacity**
  - **Diffusion Coefficients**
  - **Viscosity**
  - **Thermal Conductivity**
  - **Electrical Conductivity**



Reference:: 1. N. Ohtori, M. Salanne, and P. Madden. J. Chem. Phys. 130, 104507 (2009).
2. M. Salanne, C. Simon, P. Turq, and P. Madden, *Journal of Fluorine Chemistry* 130 (2009) 38-44.

Density Calculations